

## CLAIMS

1. A manufacturing method of a semiconductor device comprising the steps of:

forming a film containing a metal atom and a silicon atom on a substrate; and

performing a nitriding process for the film,

wherein the film is formed by changing a silicon concentration in at least two stages in the step of forming the film.

2. The manufacturing method of a semiconductor device according to claim 1, wherein the film having different silicon concentration in a depth direction is formed in the step of forming the film.

3. The manufacturing method of a semiconductor device according to claim 1, wherein the film composed of two or more layers having different silicon concentration respectively is formed in the step of forming the film.

4. The manufacturing method of a semiconductor device according to claim 1, wherein the film is formed in the step of forming the film, so that the surface side of the film has a larger silicon concentration than that of a substrate side.

5. The manufacturing method of a semiconductor device according to claim 1, wherein the film is formed in the step of forming the film, so that a surface side of

the film becomes silicon-rich, and a substrate side of the film becomes metal-rich.

6. The manufacturing method of a semiconductor device according to claim 1, wherein the film is formed by using a first source containing a metal atom and a second source containing a silicon atom, intermittently feeding each source to the substrate, and changing a supply flow rate or supply time of each source respectively.

7. The manufacturing method of a semiconductor device according to claim 1, wherein the metal atom is hafnium and the film is a hafnium silicate film.

8. The manufacturing method of a semiconductor device according to claim 1, wherein the step of performing the nitriding process is conducted in the same reaction chamber as the reaction chamber where the step of forming the film is conducted.

9. A manufacturing method of a semiconductor device, comprising the steps of:

forming a film containing a metal atom and a silicon atom on a substrate; and

introducing nitrogen into the film,

wherein a nitrogen concentration introduced into the film in the step of introducing nitrogen is controlled by a silicon concentration in the film formed in the step of forming the film.

10. A manufacturing method of a semiconductor device comprising the steps of:

carrying a substrate into a reaction chamber;

processing the substrate by feeding a first source gas obtained by vaporizing a first source which is prepared by mixing plural kinds of liquid sources, and a second source gas obtained by vaporizing a second source which is prepared by mixing plural kinds of liquid sources at a mixing ratio different from that of the first source, or composed of one kind of liquid source; and

carrying the substrate after processing out of the reaction chamber.

11. The manufacturing method of a semiconductor device according to claim 10, wherein a supply flow rate of the first source and/or second source is changed respectively in the step of processing the substrate.

12. The manufacturing method of a semiconductor device according to claim 10, wherein, the first source gas and the second source gas are simultaneously fed to the substrate in the step of processing a substrate.

13. The manufacturing method of a semiconductor device according to claim 10, wherein, the first source gas and the second source gas are alternately fed to the substrate in the step of processing the substrate.

14. The manufacturing method of a semiconductor

device according to claim 10, wherein feeding of the first source gas and the second source gas, and feeding of a third source gas different from the first and second source gases are alternately performed by at least more than once in the step of processing a substrate.

15. The manufacturing method of a semiconductor device according to claim 10, wherein feeding of the first source gas and feeding of the second source gas are alternately performed by at least more than once, with feeding of a third source gas different from the first and second source gases intervened between the feeding of the first source gas and the feeding of the second source gas.

16. The manufacturing method of a semiconductor device according to claim 10, wherein the plural kinds of liquid sources constituting the first source are a Hf liquid source and a Si liquid source, the one kind of liquid source constituting the second source is either of the Hf liquid source or the Si liquid source, and a process means to form a Hf silicate film.

17. The manufacturing method of a semiconductor device according to claim 16, wherein a mixing ratio of the Si liquid source and the Hf liquid source in the first source (Si liquid source/Hf liquid source) is set to be in the range of from 100 to 1000.

18. The manufacturing method of a semiconductor

device according to claim 16, wherein a composition ratio  $Hf/(Hf + Si)$  of the Hf silicate film formed on the substrate is controlled in a depth direction in the range of from 0.1 to 1.0, by changing a supply flow rate of the first source and/or second source in the step of processing the substrate.

19. The manufacturing method of a semiconductor device according to claim 18, further comprising the step of performing a nitriding process for the Hf silicate film formed in the step of processing the substrate.

20. A substrate processing apparatus, comprising:

a reaction chamber for processing a substrate;

a first tank for housing a first source prepared by mixing plural kinds of liquid sources;

a second tank for housing a second source prepared by mixing the plural kinds of liquid sources at a mixing ratio different from that of the first source, or composed of one kind of liquid source;

a first liquid flow rate controller that controls a liquid flow rate of the first source;

a second liquid flow rate controller that controls the liquid flow rate of the second source;

a first vaporizer that vaporizes a flow rate-controlled first source;

a second vaporizer that vaporizes a flow rate-

controlled second source; and

a feed port that feeds the first source gas and second source gas obtained by vaporization, to the reaction chamber.

21. The substrate processing apparatus according to claim 20, comprising a controller that controls to change a set value of the first liquid flow rate controller and/or the second liquid flow rate controller during processing the substrate.